LA-UR-21-28068
Approved for public release; distribution is unlimited.

Title: Evaluating New Double Perovskite Halide Scintillators for Radiation Detection Applications

Author(s): Rutstrom, Daniel Joseph
Zhuravleva, Mariya
Mcclellan, Kenneth James

Intended for: NSSC-LANL 2021 Keepin Nonproliferation Program

Issued: 2021-08-11

Evaluating New Double Perovskite Halide Scintillators for Radiation Detection Applications

Daniel Rutstrom[1,2], Ken McClellan[1], Mariya Zhuravleva[2]

Los Alamos NATIONAL LABORATORY

[1]Los Alamos National Laboratory

Materials Science and Technology (MST) Division

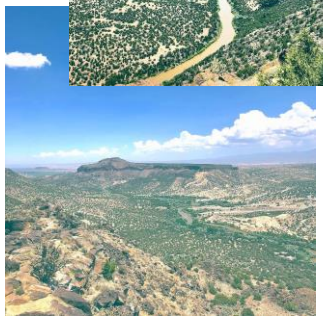
MST-8 Materials Science in Radiation and Dynamics Extremes

[2]University of Tennessee

LA-UR-XXXXXX

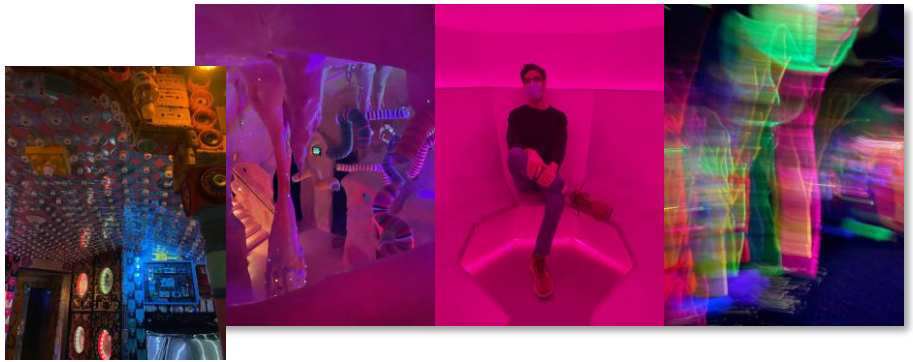
Summer Activities

White Rock Overlook



Tsankawi Trail (Los Alamos)

Meow Wolf (Santa Fe)



Daniel Rutstrom (MST-8)

- Educational Background
  - B.S. in MSE, University of Tennessee, 2018
  - M.S. in MSE, University of Tennessee, 2021

THE UNIVERSITY OF TENNESSEE KNOXVILLE

- Division
  - Materials Science in Radiation and Dynamics Extremes (MST-8)
  - Mentor: Ken McClellan

Daniel Rutstrom

Ken McClellan

- Research
  - Experimental validation of machine learning based scintillator prediction models
  - Discovery and development of advanced halide scintillators for gamma spectroscopy applications

Research Overview and Motivation

- **Objectives**
  - Provide experimental data to feed back in to models for high-throughput predictions of potential new scintillator materials
  - Identify promising scintillator compounds to pursue further

Research Overview and Motivation

- **Objectives**
  - Provide experimental data to feed back in to models for high-throughput predictions of potential new scintillator materials
  - Identify promising scintillator compounds to pursue further

- **Motivation:**
  - **Ideal scintillator does not exist, new materials with tailored properties are desired**
  - Discovery of new scintillator compounds can be lengthy and tedious
  - Complimentary use of modeling and experimental work can provide a more efficient approach than conventional "trial and error" approach

    → **Rapid screening, vast chemical spaces**

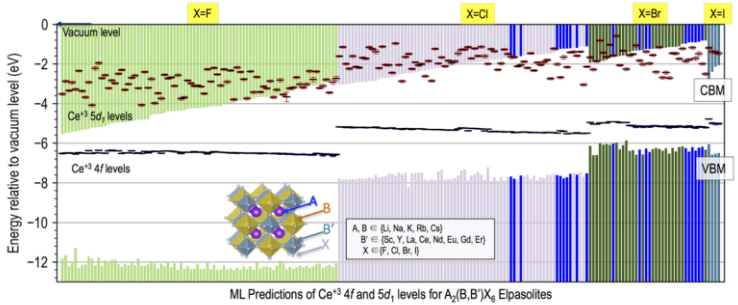
- **Applications:** gamma spectroscopy, flash radiography, etc.

Research Approach

Fabrication of predicted new scintillators

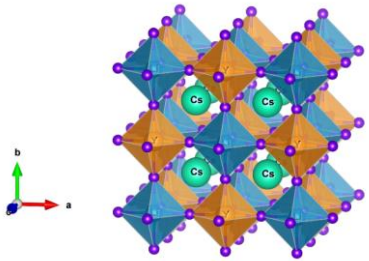
- **Selected a set of new compounds to pursue experimentally, based on calculated electronic band structures**
- **Compositional space – double perovskite halides** → $A_2BB'X_6$:Ce (A = Rb^+, Cs^+ ; B = Na^+, K^+ ; B' = RE^{3+} ; X = Cl^-, Br^-)
- **Fabrication method – single crystal growth via Bridgman technique**
- **Characterization – physical, optical, and scintillation properties**

Positions of Ce^{3+} 5*d* and 4*f* energy levels relative to CB and VB for 200 Double Perovskite Halides



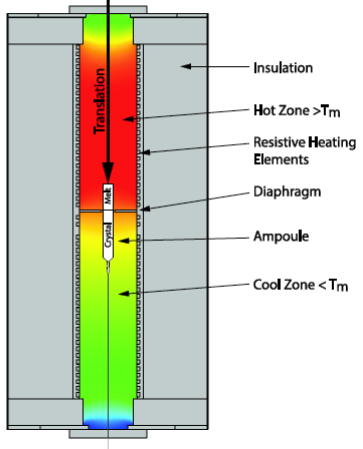
G. Pilania *et al.* "Physics-informed machine learning for inorganic scintillator discovery" J. Chem. Phys. **148**, 241729 (2018)

Double perovskite structural model (Cs_2LiYCl_6)



Cubic space group *Fm*-3*m*

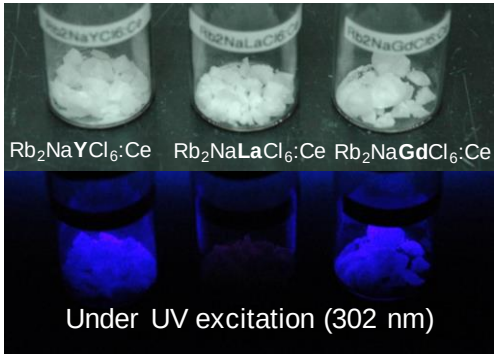
Bridgman Method



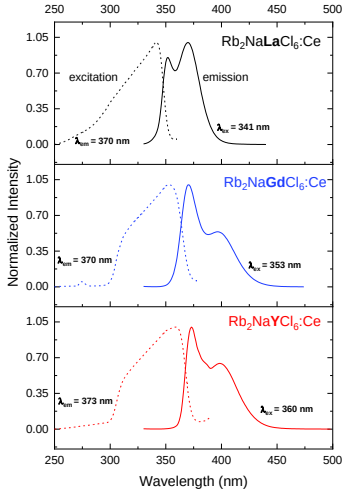
Summary of Results

- **Synthesized 3 new compounds**
  - Rb_2NaYCl_6:Ce
  - $Rb_2NaLaCl_6$:Ce
  - $Rb_2NaGdCl_6$:Ce
- **All three compositions luminesce**
- **$Rb_2NaGdCl_6$:Ce is most promising, highest intensity emission**
- **Determined melting points via DSC** (not reported in literature since these are new materials)
- **In progress**
  - Crystal growth
  - X-ray diffraction

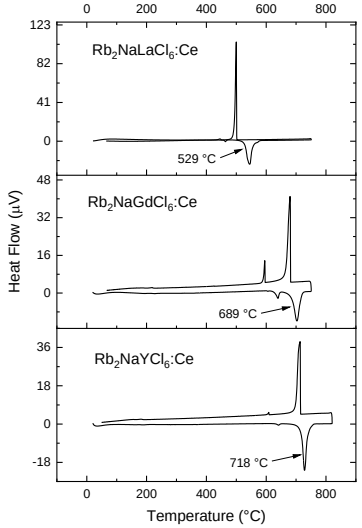
Synthesized polycrystalline samples



Photoluminescence



Differential Scanning Calorimetry (DSC)



Radioluminescence (X-ray) Emission Spectra